

**Supplementary Information: Fluoride Removal by Calcite and Fluoroapatite**

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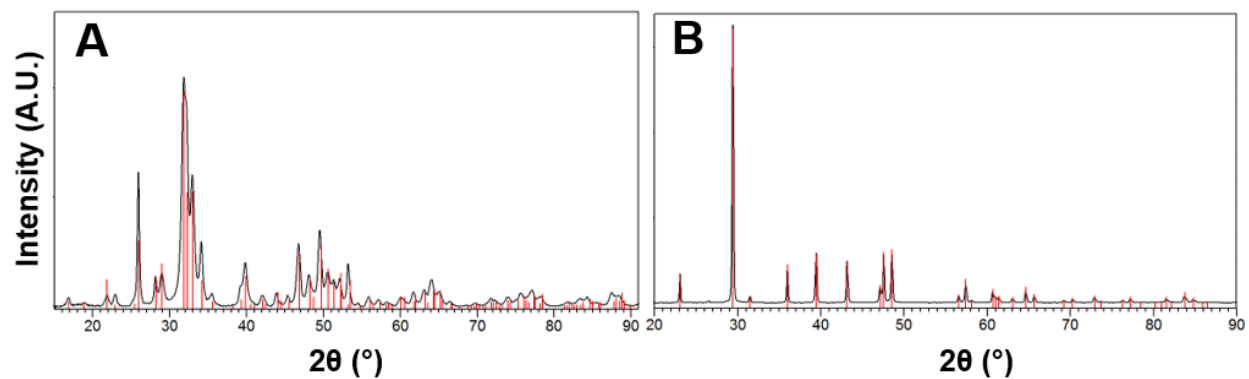
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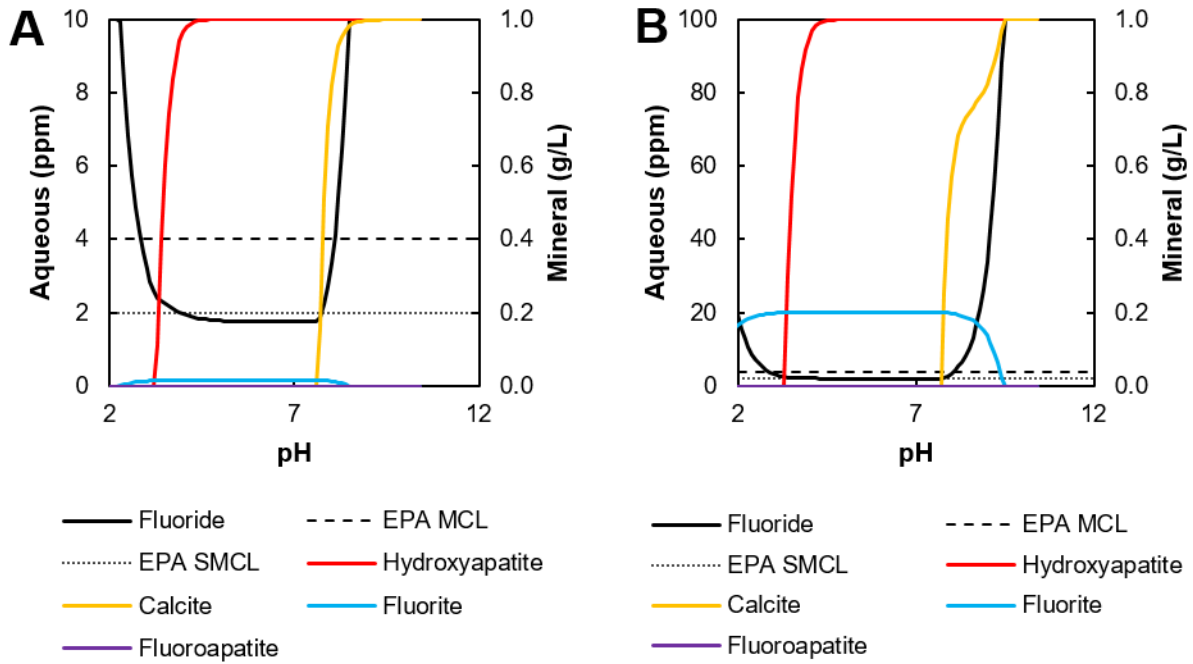
Table S1. Synthetic groundwater compositions.<sup>1,2</sup>

<b>Ion</b>	<b>Concentration (ppm)</b>			
	<b>Region</b>	<b>Ghana</b>	<b>Tanzania</b>	<b>Sri Lanka</b>
<b>Ca</b>		27.6	17.6	173
<b>Mg</b>		13.7	1.40	179
<b>HCO<sub>3</sub></b>		146	845	516
<b>SO<sub>4</sub></b>		1.70	20.8	15.5
<b>Si</b>		34.4	54.2	45.0
<b>NO<sub>3</sub> as N</b>		4.90	7.80	9.00
<b>Ionic strength</b>		0.012	0.028	0.042

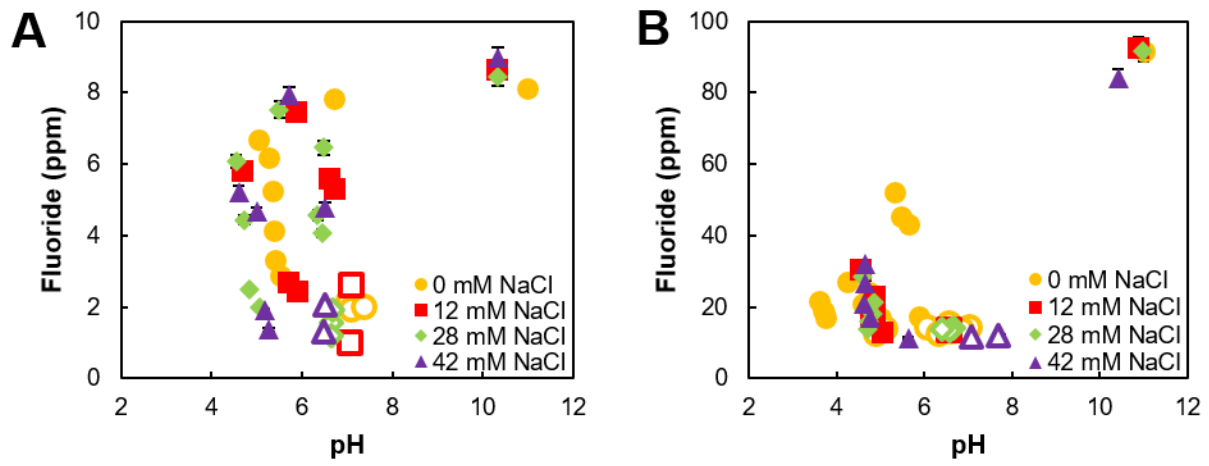
**Figure S1.** XRD patterns of the starting A) hydroxyapatite and B) calcite materials. Black lines are the data; red lines show JCPDS reference cards 01-075-0425 (A) and 01-083-0578 (B).



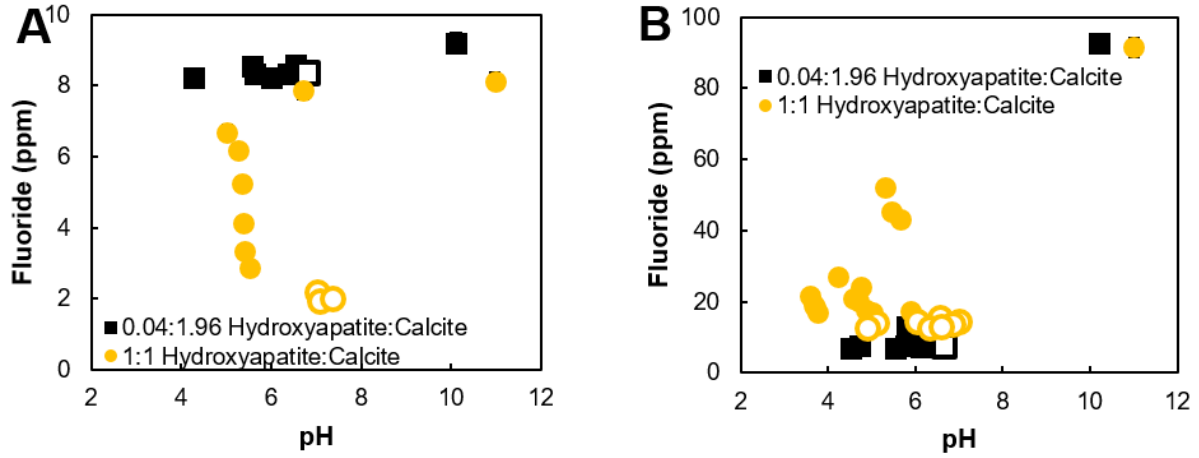
**Figure S2.** Thermodynamic modeling of fluoride sequestration at 10 ppm (A) and 100 ppm (B). Initial hydroxyapatite and calcite suspension densities were both 1 g/L. No background electrolyte was added.



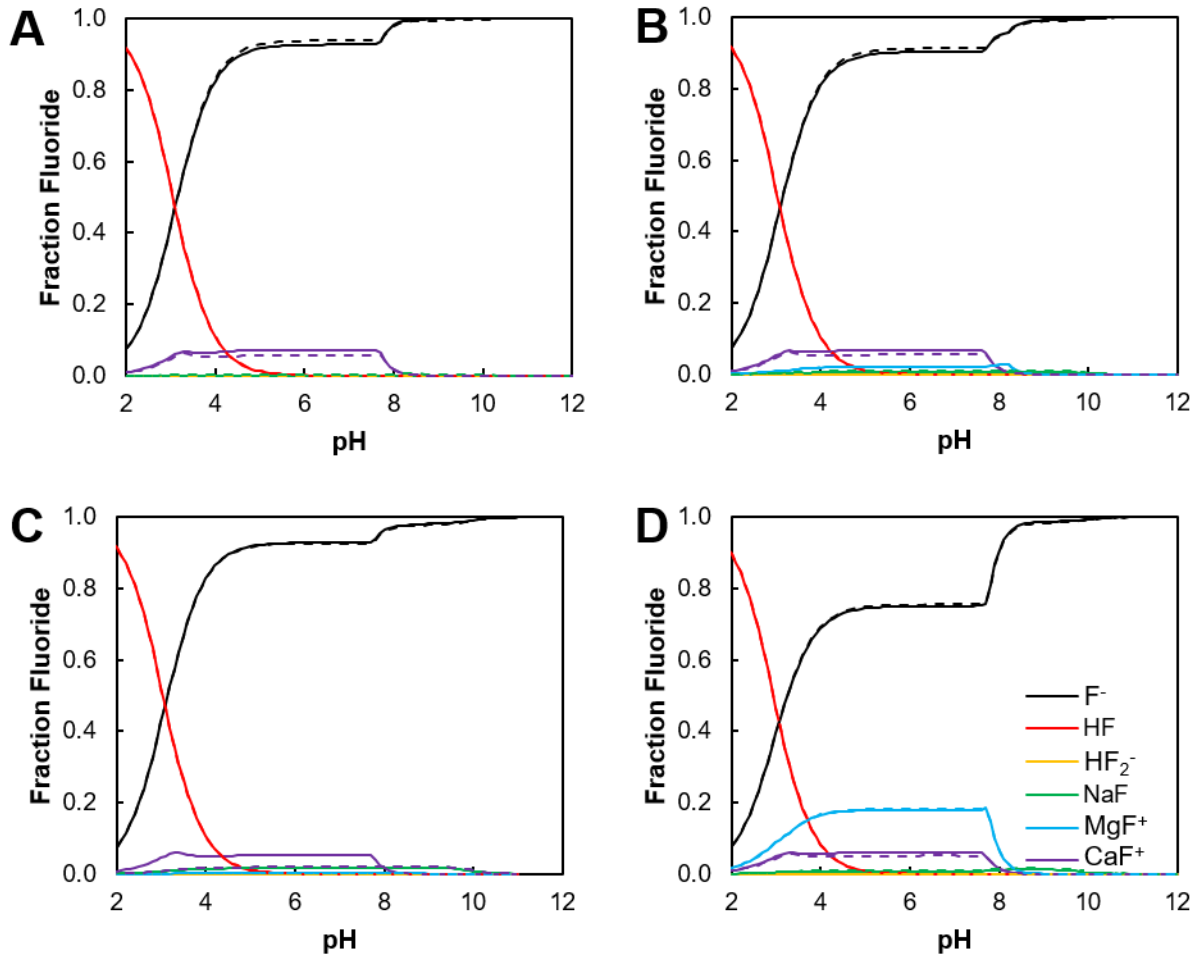
**Figure S3.** Effects of ionic strength on fluoride sequestration at 10 ppm fluoride (A) and 100 ppm fluoride (B). Closed symbols indicate acidification steps, and open symbols indicate neutralization steps.



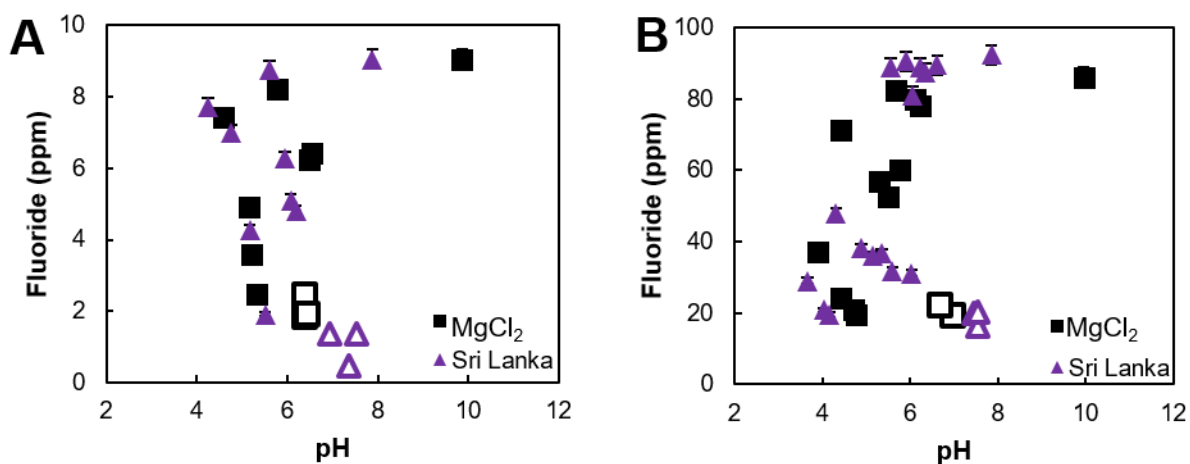
**Figure S4.** Fluoride removal at 10 ppm fluoride (A) and 100 ppm fluoride (B). Hydroxyapatite suspension density was 0.04 g/L, and calcite suspension density was 1.86 g/L. No background electrolyte was added. Closed symbols indicate acidification steps, and open symbols indicate neutralization steps.



**Figure S5.** Thermodynamic modeling of fluoride speciation at 10 ppm (solid lines) and 100 ppm (dashed lines) for no background electrolyte (A) and simulated Ghana (B), Tanzania (C), and Sri Lanka (D) groundwaters. Hydroxyapatite and calcite suspension densities were both 1 g/L.



**Figure S5.** Fluoride removal at 10 ppm fluoride (A) and 100 ppm fluoride (B) in the presence of 7.36 mM MgCl<sub>2</sub>. Hydroxyapatite and calcite suspension densities were both 1 g/L. Closed symbols indicate acidification steps, and open symbols indicate neutralization steps.





## References

- 1 W. M. Edmunds and P. L. Smedley, in *Essentials of Medical Geology: Revised Edition*, ed. O. Selinus, Springer Netherlands, Dordrecht, 2013, pp. 311–336.
- 2 K. Cherukumilli, C. Delaire, S. Amrose and A. J. Gadgil, Factors Governing the Performance of Bauxite for Fluoride Remediation of Groundwater, *Environ. Sci. Technol.*, 2017, **51**, 2321–2328.